

## MERCURY AND METHYLMERCURY CONCENTRATIONS IN TWO NEWLY CONSTRUCTED RESERVOIRS IN THE WUJIANG RIVER, GUIZHOU, CHINA

HENG YAO,<sup>†‡</sup> XINBIN FENG,<sup>\*†</sup> YANNA GUO,<sup>†</sup> HAIYU YAN,<sup>†</sup> XUEWU FU,<sup>†</sup> ZHONGGEN LI,<sup>†</sup> and BO MENG<sup>†‡</sup><sup>†</sup>State Key Laboratory of Environmental Geochemistry, Chinese Academy of Sciences, Guiyang, China<sup>‡</sup>Graduate University, Chinese Academy of Sciences, Beijing, China

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**Abstract**—We studied the distribution of different mercury species in two newly created reservoirs (Hongjiadu [HJD] and Suofengying [SFY] Reservoir) within the Wujiang River, Guizhou Province, China. These reservoirs were sampled four times between January, 2007, and November, 2007. Water, soil, and fish tissue samples were collected and analyzed for organic (methylmercury) and total Hg. Dissolved organic carbon (DOC) content in water was measured as well. In both reservoirs, the DOC concentration was low (range 0.5–4.9 mg/L). Organic matter content in soil collected near the bank of both reservoirs was also low (range 0.4–6.9%). The total MeHg level did not increase significantly with depth in water column, nor did it exhibit a pronounced spatial pattern moving from upstream to the dam. The total MeHg level did not increase significantly with depth in water column, nor did it exhibit a pronounced spatial pattern moving from upstream to the dam. The total Hg content in fish tissue was on average 0.044 mg/kg wet weight, which is a very low content in an international comparison. It is suggested that the studied reservoirs were not active sites of net Hg methylating. Low levels of organic matter (OM) may constrict the evolution of the bulk Hg methylation process occurring in flooded soil. Therefore, we hypothesize that newly constructed reservoirs in the karstic region of Southwest China appear to be much less beset by the problems of MeHg pollution that have been reported for corresponding reservoirs in North America and Europe. However, further research is required to verify this finding. *Environ. Toxicol. Chem.* 2011;30:530–537. © 2010 SETAC

**Keywords**—Reservoir    Methylmercury    Organic matter    Soil

## INTRODUCTION

Methylmercury (MeHg) is one of the most important forms of mercury, because it readily bioaccumulates in food chains and is a potent neurotoxin for humans and wildlife [1]. It is widely reported that Hg concentrations in predatory fish in newly constructed reservoirs in North America and Europe exceeded 1.0 mg/kg [2–8], the consumption limit of MeHg in fish recommended by the Joint Food and Agriculture Organization of the United Nations/World Health Organization Expert Committee on Food Additives [9]. The high levels of MeHg persist for decades after initial flooding [10,11]. The decomposition of organic matter (OM) from flooded soils and increased microbial methylation of inorganic Hg<sup>II</sup> to MeHg species after flooding are believed to be responsible for the elevation of MeHg in fish [12–15]. However, the factors that impact Hg pollution in newly created reservoirs deserve further study.

China has constructed the largest number of reservoirs in the world (86,353 in total), including 17,126 reservoirs created in southwestern China, accounting for 20% of the total in China [16]. However, studies on Hg biogeochemical cycling in newly built reservoirs in China are limited. Feng et al. [17] investigated the biogeochemical cycling of Hg in two relatively old reservoirs created in the Wujiang River, Guizhou, China, and reported that a high primary productivity in the reservoir generated elevated levels of OM in sediment that in turn enhanced MeHg production. However, the status of MeHg contamination in newly constructed reservoirs in China is still unknown.

The Wujiang River is the largest upstream tributary of the Yangtze River, located in the Yunnan-Guizhou plateau in

southwestern China. Eleven cascade hydroelectric power stations have been planned and built along the Wujiang River, which are expected to alter its aquatic environment. The newly built reservoirs may become sites more suitable for the methylation and bioaccumulation of MeHg in the aquatic food chain [10]. Many more reservoirs have been or will be built in this region on the Zujiang River, Beipanjiang River, Nanpanjiang River, and Jinshajiang River. All of these rivers have a geological background (limestone as the major bedrock), geographical environment, hydrological characteristics, and climatic condition similar to those of the Wujiang River. Therefore, the outcome of the present study in the Wujiang River can be used to predict MeHg contamination status of a large number of reservoirs in southwestern China.

In the present study, we chose two newly created reservoirs within the Wujiang River as the study area. The Hongjiadu (HJD) and Suofengying (SFY) reservoirs, built in 2004 and 2005, respectively, are located upstream of the Wujiang River in Guizhou Province, China. These two newly built reservoirs provide a unique opportunity to investigate the behavior of Hg in aquatic systems and to understand the mechanisms of MeHg production in newly built reservoirs. The present study reports the distribution and speciation of Hg in the water column and total Hg concentration in fish tissue in these two reservoirs. We also measured OM content in soil along the banks of both reservoirs to represent OM contents in submerged soil of both reservoirs.

## MATERIALS AND METHODS

*Sampling sites description*

The basic characteristics of HJD and SFY reservoirs are listed in Table 1. Hongjiadu is the largest reservoir of the 11 cascade reservoirs created in Wujiang River, whereas Suofengying is the smallest. The predominant inflow into HJD Reservoir is from the Liuchong and Aoshui Rivers. The inflows to

\* To whom correspondence may be addressed  
(fengxinbin@vip.skleg.cn).

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Table 1. Hydrological and basic parameters of the Hongjiadu and Suofengying reservoirs, China

|                          | Unit                    | Hongjiadu       | Suofengying     |
|--------------------------|-------------------------|-----------------|-----------------|
| Year of construction     |                         | 2004            | 2005            |
| Total water volume       | $10^8 \text{ m}^3$      | 49.47           | 2.01            |
| Watershed area           | $\text{km}^2$           | 80.5            | 5.7             |
| Maximum depth            | m                       | 110             | 80              |
| Mean depth               | m                       | 61.5            | 35.5            |
| Water residence time     | d                       | 380             | 7               |
| Average annual flow      | $\text{m}^3/\text{sec}$ | 155             | 395             |
| pH                       |                         | $7.6 \pm 0.4$   | $7.8 \pm 0.3$   |
| Dissolved oxygen         | mg/L                    | $5.4 \pm 2.1$   | $7.5 \pm 1.6$   |
| Dissolved organic carbon | mg/L                    | $0.62 \pm 0.17$ | $0.68 \pm 0.18$ |
|                          |                         | (winter)        | (winter)        |
|                          |                         | $2.10 \pm 0.51$ | $3.45 \pm 0.73$ |
|                          |                         | (spring)        | (spring)        |
|                          |                         | $1.35 \pm 0.54$ | $2.35 \pm 0.96$ |
|                          |                         | (summer)        | (summer)        |
|                          |                         | $0.93 \pm 0.28$ | $1.00 \pm 0.28$ |
|                          |                         | (autumn)        | (autumn)        |

SFY include a major one from the Dongfeng Reservoir and a relatively minor one from the Maotiao River. Besides the influx from rivers, runoffs from the upland seasonally contribute to the reservoirs. The reservoirs experience a typical subtropical humid monsoon climate. The rainy season occurs in the spring and summer seasons accounting for more than 70% of the annual precipitation. The reservoirs were created for a variety of purposes, including the production of hydroelectricity, irrigation, flood control, fisheries production, and recreation. These reservoirs are located in rural areas, remote from industrial activities and densely populated urban centers. Seventy-two percent of the flooded area is composed of former agricultural lands with an underlying bedrock of limestone and dolomite.

#### Sampling and analytical methods

Water samples were collected on a seasonal basis during 2007 at stations representing the upstream (A,E), midstream (B,F), and approximate to dam (C,G) sections of the reservoirs and also within the tributaries (D,H). In both of the reservoirs, the sampling sites were oriented in a west-to-east direction, following the water flow (Fig. 1). Seasonal vertical column profiles were established at each site, A to H, by sampling at eight depths (0, 5, 10, 20, 40, 60, 80, 100 m). A 5-L Niskin



Fig. 1. Location of the sampling sites near the Hongjiadu and Suofengying reservoirs within the Wujiang River, Guizhou Province, China.

bottle water sampler manufactured from polytetrafluoroethylene (model 1010; General Oceanics) was employed to collect in total 189 water samples at various depths. The Niskin bottle was during sampling positioned in the upstream direction relative to the operator.

Monthly integrated bulk precipitation was collected by using an in-house-built sampler, following the design recommended by the Joint Assessment and Monitoring Programme of the Operational and Strategic Planning and Research Commission [18]. After collection, the precipitation sample was stored in a precleaned 800-ml borosilicate bottle including 10 ml concentrated HCl with low Hg blank, which was added to prevent adsorption of Hg to the surface of the container. Precipitation was collected on a monthly basis during all of 2006 [19].

Following a cleaning protocol for trace metal analysis, borosilicate glass bottles were cleaned in dilute acid (10%  $\text{HNO}_3$ ), rinsed three times with ultrapure deionized water ( $18.2 \text{ M}\Omega \cdot \text{cm}$ ), then heated in a muffle furnace to  $500^\circ\text{C}$  and were, after cooling, double bagged and stored in plastic boxes before sampling [20]. Water samples were collected into borosilicate glass bottles immediately at the sampling site. All water samples were acidified with 0.5% (v/v) concentrated ultrapure hydrochloric acid as well as double bagging on site and were after transportation stored at  $4^\circ\text{C}$  in the dark until analyzed [20]. Every water sample was analyzed for total Hg (THg, unfiltered) as well as total methylmercury (TMeHg, unfiltered). Concerning the determination of THg, a sample was initially oxidized by 0.5%  $\text{BrCl}(\text{aq})$  for 24 h, and subsequently the free halogens remaining behind were titrated by  $\text{NH}_2\text{OH} \cdot \text{HCl}(\text{aq})$  before adding  $\text{SnCl}_2(\text{aq})$  to convert  $\text{Hg}^{\text{II}}$  to volatile  $\text{Hg}^0$ . The resulting sample was purged of  $\text{Hg}^0(\text{aq})$  with Hg-free  $\text{N}_2$ , then the analytic was quantified using the dual-stage gold amalgamation method coupled with a cold vapor fluorescence (CVAFS) detector (Tekran model 2500 instrument; Tekran) [20,21]. Total methylmercury in water samples was analyzed using distillation, ethylation, and GC-CVAFS detection, following United States Environmental Protection Agency (U.S. EPA) method 1630. A 45-ml aliquot portion was distilled at  $125^\circ\text{C}$  in a Teflon vessel until approximately 35 ml distillate was received. The sample collected was transferred into a 150-ml bubbler after adding 0.2 ml 2 M sodium acetate buffer and 100  $\mu\text{l}$  of the ethylation agent  $\text{NaB}(\text{C}_2\text{H}_5)_4(\text{aq})$  (1% solution) and then left to react at room temperature for 15 min. Volatile peralkylated products,  $\text{CH}_3\text{HgC}_2\text{H}_5$  and  $(\text{C}_2\text{H}_5)_2\text{Hg}$ , were purged from solution in a stream of  $\text{N}_2$  onto a Tenax trap. The trapped organomercurials were thermally desorbed into an argon gas stream and separated chromatographically before being thermodecomposed into  $\text{Hg}^0$ , detected with a CVAFS instrument (model III; Brooks Rand Labs) [22]. Dissolved oxygen (DO) and pH of water samples were measured in situ by a portable analyzer (PB-5010), whereas the corresponding dissolved organic carbon (DOC) content was quantified in the laboratory by using a high-temperature combustion technique [23].

Several specimens of fresh-caught wild fish species were obtained from the HJD Reservoir. The weight and length of a specimen were recorded before a sample of the dorsal muscle was isolated, bagged, and deep-frozen prior to THg analysis. A fish sample was digested in a nitrosulfuric acid mixture,  $\text{HNO}_3:\text{H}_2\text{SO}_4$  (7:3 v/v), at  $95^\circ\text{C}$  for 3 h and subsequently diluted to be processed as described above for a THg water sample [24].

Surface soil samples were in both reservoirs collected along the brink of water in order to represent flooded soil. Figure 1 shows

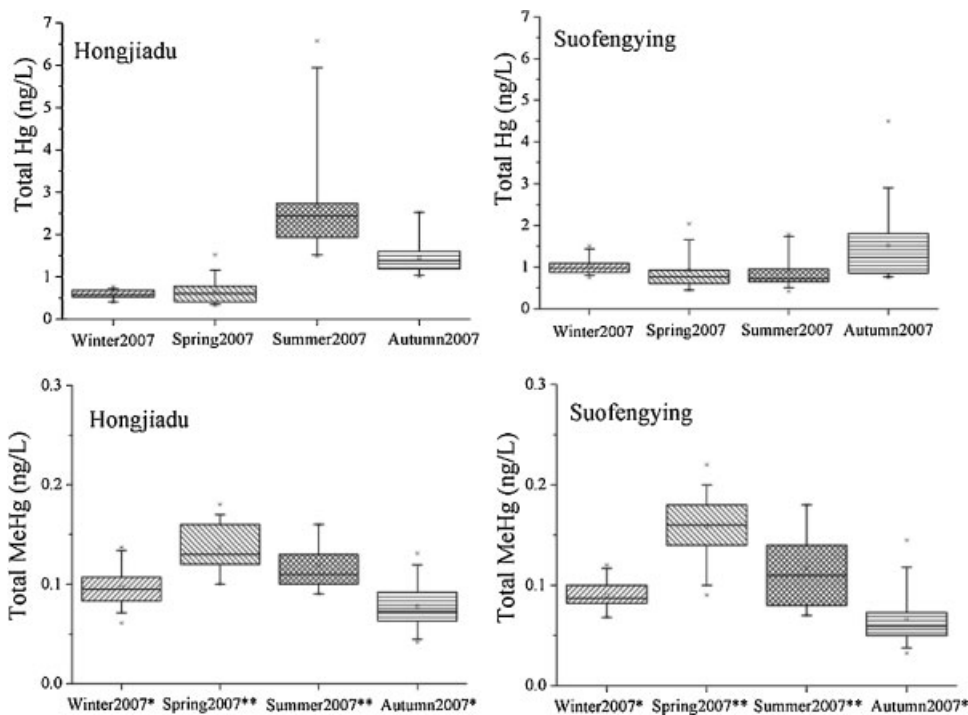


Fig. 2 Seasonal variability of total mercury (Hg) and methylmercury (MeHg) concentrations (ng/L) at different seasons in the water column in the Hongjiadu and Suofengying reservoirs, China. Each box and whisker plot shows the median, the interquartile range (25th to 75th percentile; box), and the minimum and maximum concentrations (whisker) at a specific sampling session. Total MeHg concentrations in spring and summer were significantly higher ( $p < 0.05$ ,  $**p < 0.001$ ) than in winter and autumn.

pairs of soil and water sampling sites from the water column. All samples were stored in acid-cleaned high-density polyethylene centrifuge tubes and were transported to the laboratory and stored frozen. Soil samples were freeze-dried and homogenized with mortar and pestle, then sieved through a 80- $\mu\text{m}$  sieve to remove coarse particles and biologic debris before THg analysis. The determination was initiated by acid digestion ( $\text{HCl}:\text{HNO}_3 = 1:3$  v/v) followed by the CVAFS detection method [25]. The concentration of organic matter in the soil samples was analyzed by using  $\text{K}_2\text{Cr}_2\text{O}_7$  oxidation coupled with volumetric technique [26].

#### Quality control

Quality control was exercised using duplicates, method blanks, blank spikes and matrix spikes, and standard reference material. Blank spikes and duplicates were taken regularly ( $>10\%$  of samples) throughout each sampling campaign. The method detection limit (MDL), based on three times the standard deviation of replicate measurements of a blank solution in the water sample, was as follows: 0.20 ng/L for THg, 0.032 ng/L

for TMeHg, respectively, and the method blank was found to be less than the detection limits in all cases. The average relative standard deviations for the duplicate analysis of THg and TMeHg for water samples were from 2.5 to 12.6% and from 4.4 to 7.5%, respectively. Spike recoveries for THg and TMeHg in water samples were between 90 and 110%, and between 84.1 and 113.1%, respectively. The mean THg concentration of standard materials of GBW07305 and GBW07405 was  $0.10 \pm 0.10$  and  $0.30 \pm 0.08$  mg/kg, which is comparable with certified value of  $0.10 \pm 0.02$  and  $0.29 \pm 0.04$  mg/kg. A fish standard reference material, Lobster Hepatopancreas Reference Material for Trace Metals (NRCC-TORT-2; [http://www.nrc-cnrc.gc.ca/obj/inms-ienm/doc/crm-mrc/eng/TORT-2\\_e.pdf](http://www.nrc-cnrc.gc.ca/obj/inms-ienm/doc/crm-mrc/eng/TORT-2_e.pdf)), was also analyzed. Total Hg average concentration was  $280 \pm 20$  ng/g, which is comparable to the certified value, which is  $270 \pm 60$  ng/g.

#### Statistical analysis

The assumption of the parametric procedure was examined using one-sample K-S test, and the result showed that the data

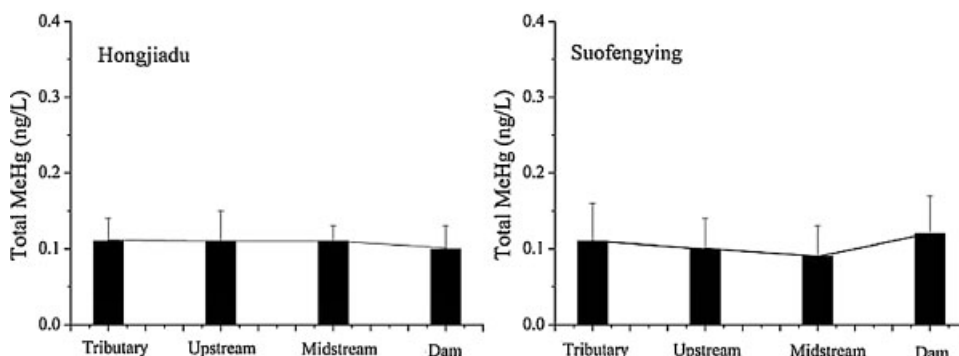


Fig. 3. Average concentration of total methylmercury (MeHg) from upstream to the dam in the Hongjiadu and Suofengying reservoirs, China. Total MeHg concentrations did not show a significant change from upstream to downstream in either reservoir ( $p > 0.05$ ), and no statistical differences were observed for total MeHg concentrations between Hongjiadu and Suofengying reservoirs ( $p > 0.05$ ).

sets followed a normal distribution; then, we used one-way analysis of variance (ANOVA) and multiple comparison to analyze the significance of differences in concentrations (e.g., TMeHg levels) among seasons and sites. Pearson's values were used as a guide to determine significance at the 5% level. Linear regression and ANOVA were performed with R. All statistical analyses were performed in the software Origin 8.0 (OriginLab) and SPSS 16.0 (SPSS).

## RESULTS AND DISCUSSION

### Mercury distribution in flooded soil

We did not manage to collect any samples of flooded soil; however, as proxies we collected numerous soil samples along the banks of the reservoirs to represent the submerged soil. Total Hg concentrations ranged from 20.1 to 133 ng/g, with a mean ( $\pm$ SD) concentration of  $68.8 \pm 34.5$  ng/g ( $n = 25$ ) in soil

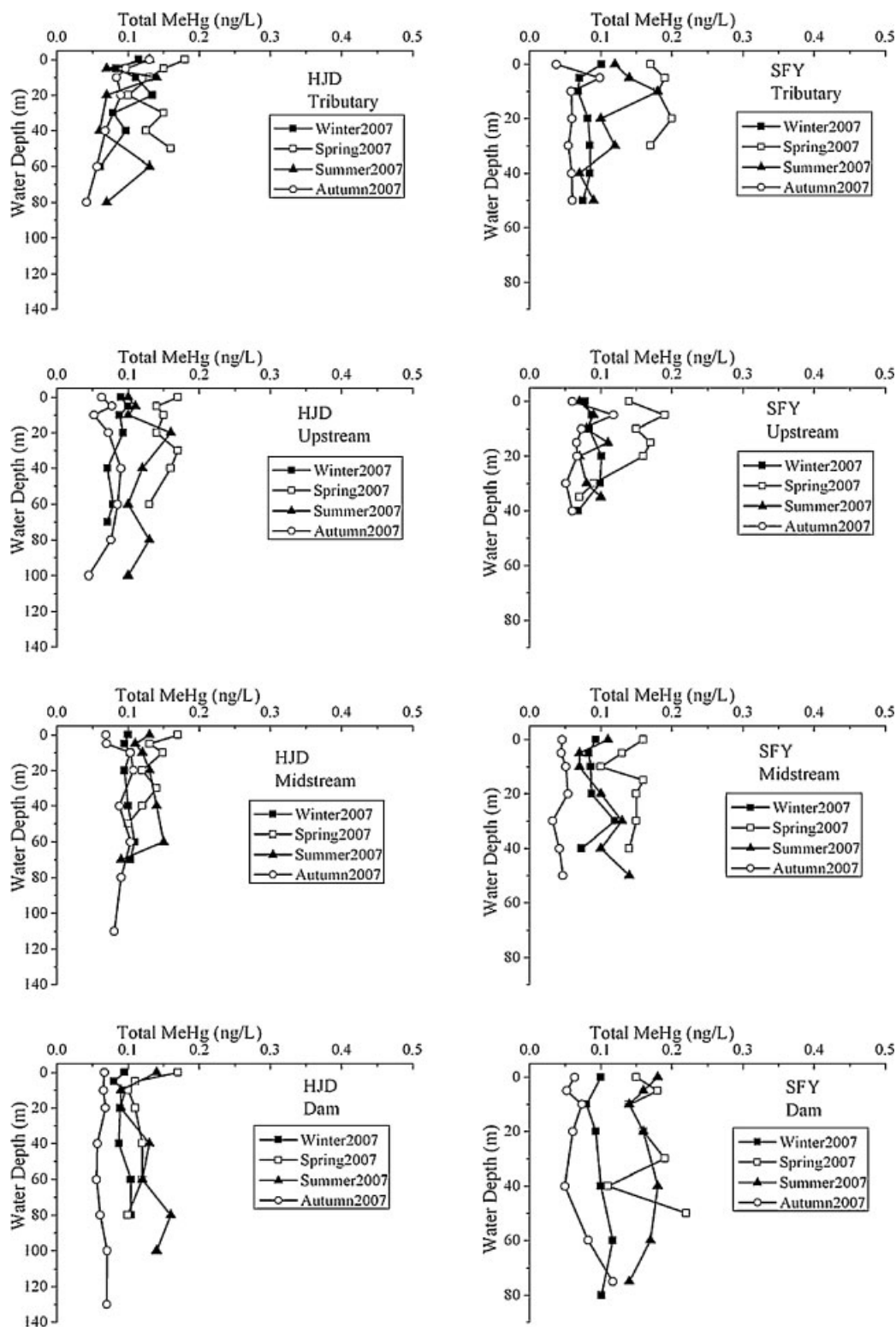


Fig. 4. Distribution of total methylmercury (MeHg) at different sampling sites in four seasons in the Hongjiadu and Suofengying reservoirs, China.

Table 2. Hg concentrations (ng/L) and total methylmercury (TMeHg)/total mercury (THg) (%MeHg) upstream and at the dam from Hongjiadu (HJD) and Suofengying (SFY) reservoirs in the Wujiang River

| Sampling sites | Reservoir   |             |              |             |             |              |
|----------------|-------------|-------------|--------------|-------------|-------------|--------------|
|                | Upstream    |             |              | Dam         |             |              |
|                | TMeHg       | THg         | %MeHg        | TMeHg       | THg         | %MeHg        |
| HJD            | 0.10 ± 0.04 | 1.61 ± 1.63 | 11.82 ± 9.31 | 0.10 ± 0.03 | 1.29 ± 0.70 | 9.12 ± 5.50  |
| SFY            | 0.11 ± 0.04 | 0.79 ± 0.23 | 13.73 ± 8.78 | 0.12 ± 0.04 | 1.20 ± 0.56 | 12.89 ± 8.15 |

Table 3. Mass balance of total mercury (THg) and total methyl mercury (TMeHg) for Hongjiadu (HJD) and Suofengying (SFY) reservoirs, China (g/y)<sup>a</sup>

| Sampling sites | Total inputs |         | Total outputs |       | Net fluxes (∑input-output) |        |
|----------------|--------------|---------|---------------|-------|----------------------------|--------|
|                | THg          | TMeHg   | THg           | TMeHg | THg                        | TMeHg  |
| HJD            | 20,397       | 506.1   | 8,794         | 432.2 | -11,603                    | -73.9  |
| SFY            | 14,307       | 1,008.3 | 12,693        | 827.7 | -1,614                     | -180.6 |

<sup>a</sup> A positive result indicates that these reservoirs are a source of TMeHg or THg and a negative result indicates that these reservoirs are a sink for TMeHg or THg [19].

samples collected along the bank of the HJD Reservoir, and the THg concentrations in soil samples collected along the bank of the SFY Reservoir ranged from 141 to 372 ng/g, with a mean (±SD) concentration of 268 ± 72.9 ng/g ( $n = 19$ ). As a result, THg concentrations in soil along the bank of the SFY Reservoir were significantly higher ( $p < 0.05$ ) than in the HJD Reservoir, and the SFY Reservoir is located downstream from an Hg mining area, as shown in Figure 1. Therefore, this is not a typical background site but rather one directly influenced by nearby Hg mining activity. Overall, the background THg concentrations of soil samples in these two reservoirs (especially SFY) are much higher than the background concentration of Hg in soil in China, which is 38.0 ng/g [27]. Moreover, we found that THg concentrations in precipitation (HJD, 41.2 ± 24.1 ng/L; SFY, 51.6 ± 38.4 ng/L) were much higher than those reported in relatively pristine areas in North America and Europe, which was generally lower than 10.0 ng/L [1,14,28]. The enhancement is attributed to regional Hg pollution by coal combustion. On the contrary, the volume-weighted mean concentrations of MeHg (±SD) in precipitation collected at HJD and SFY Reservoirs were merely 0.10 ± 0.05 ng/L and 0.12 ± 0.06 ng/L, respectively [20], which is a low level in the same reference framework. Therefore, the contribution of MeHg from precipitation to the studied reservoirs could be considered trivial in a comparison to the impact from other sources.

#### THg and TMeHg concentrations in the water column

Figure 2 displays the seasonal variability of THg in the water column in the HJD and SFY reservoirs. In the HJD Reservoir, THg concentrations decreased in the following order: summer > autumn > spring and winter. Corresponding to the increase in rainfall during the summer and autumn, abundant surface water carried more particulate matter with higher THg concentrations to the HJD Reservoir. In the SFY Reservoir, THg levels were lower than those in HJD, possibly as a result of more extensive settling of particles in the Dongfeng Reservoir, which is located upstream of the SFY Reservoir.

The seasonal variability in TMeHg in the water column of both reservoirs is shown in the lower panel of Figure 2. In general, in the spring and summer TMeHg concentration was significantly higher ( $p < 0.001$ ) than that during the winter and autumn. In both reservoirs, we found that DOC was positively correlated with TMeHg in the water column (HJD,  $R^2 = 0.13$ ,  $p < 0.0001$ ,  $n = 116$ ; SFY,  $R^2 = 0.31$ ,  $p < 0.0001$ ,  $n = 106$ ). DOC levels (Table 1) were not as high as those observed in peat bog lakes in North America, where DOC concentration reached up to 11 mg/L [10,29]. Table 1 shows that DOC concentrations in the spring and summer were higher than those in the autumn and winter, which may be due to transportation of DOC from soil by soil erosion induced by precipitation. Therefore, we suggest that the low TMeHg concentrations in both reservoirs were associated with low DOC in the water column [30,31].

No statistical difference ( $p > 0.05$ ) could be established between the mean TMeHg concentration of the HJD and SFY reservoirs. From the vertical concentration–depth profiles shown in Figure 3, it is obvious that TMeHg did not increase significantly with depth in the water column. Analogously, no discernible change in TMeHg could be detected from upstream to the dam moving along the sampling section of the reservoirs (Fig. 4), and the mean MeHg concentrations at the site close to the dam were consistent with the values observed upstream in both reservoirs (Table 2). This demonstrated that these two newly built reservoirs did not show net Hg methylation in the aquatic systems. The percentage of THg that occurs as MeHg

Table 4. Comparison of total mercury (THg) and total methylmercury (TMeHg) concentrations in the water columns in Hongjiadu and Suofengying reservoirs, China, with reservoirs in North America and Europe (ng/L)

| Sampling location                 | Periods | THg (ng/L)          | TMeHg (ng/L)            | Data source                |
|-----------------------------------|---------|---------------------|-------------------------|----------------------------|
| Hongjiadu Reservoir, China        | 4       | 0.3–6.6 (1.4 ± 1.1) | 0.05–0.17 (0.11 ± 0.03) | Present study              |
| Suofengying Reservoir, China      | 3       | 0.4–4.9 (1.3 ± 0.7) | 0.03–0.22 (0.11 ± 0.04) | Present study              |
| Southern Illinois, USA            | 5       | <10                 |                         | Cox et al. [3]             |
| ELA, Northwestern Ontario, Canada | 0.08    | 4.5 ± 1.7           | 1.4 ± 0.5               | Hall et al. [14] Rudd [32] |
|                                   | 3       | 1.1–13.3            | 0.19–2.66               |                            |
| Northern Québec, Canada           | 3       | <10                 | 0.01–2                  | Lucotte et al. [10]        |
| Northwestern Minnesota, USA       | 4       | 0.74–6.97           | 0.056–6.6               | Brigham et al. [40]        |
| Southern and central Finland      | 5       | 4.8 ± 2.3           | 0.77 ± 0.61             | Porvari and Verta [41]     |

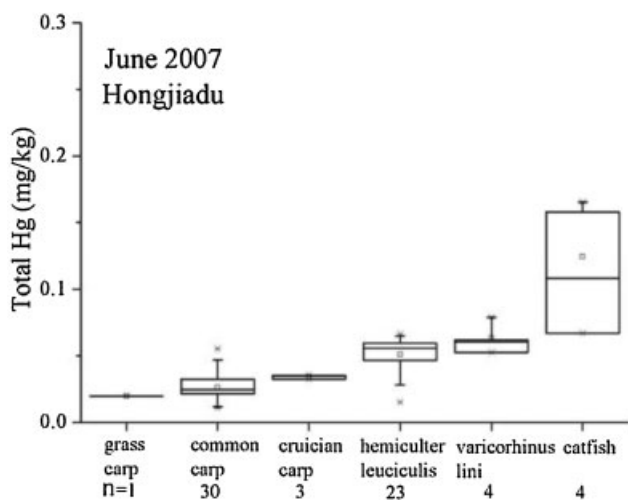


Fig. 5. Total Hg concentrations (in mg/kg) measured in different fish species collected from the Hongjiadu Reservoir, China. Box and whisker plot shows the median, the interquartile range (25th to 75th percentile; box), and the minimum and maximum concentrations (whisker).

(%MeHg) is a good indicator of MeHg production in ecosystems [32–34]. Therefore, we calculated percentage MeHg ratios in the HJD and SFY reservoirs. The percentage MeHg at the sites close to the dam was somewhat lower than that at the upstream sites in both reservoirs (Table 2), and the percentage MeHg in both reservoirs was much lower than the percentages in newly built reservoirs reported in North America and Europe (60–80%) [1,14]. Moreover, Guo [19] conducted a mass balance study of both THg and MeHg based on one-year continuous measurement of monthly average THg and MeHg concentrations in inflows and outflows in these two reservoirs, which indicated that both reservoirs were net sinks for THg and TMeHg, as shown in Table 3. Therefore, our data confirmed that these two new reservoirs are not active sites for net Hg methylation.

A comparison of Hg concentrations between these two newly built reservoirs and reservoirs in other areas is shown in Table 4. Total Hg concentrations in the present study were

somewhat lower than those in reservoirs in North America and Europe, but TMeHg concentrations in the water column of both the SFY and HJD reservoirs were significantly lower than those in newly built reservoirs in North America and Europe. This also indicated that no net MeHg production occurred in either of these reservoirs, which is contrary to newly built reservoirs in Europe and North America.

#### THg in fish

We collected fish samples only in the HJD Reservoir, including predatory fish (catfish), polyphagous fish (common carp, crucian carp, *Hemiculter leuciculis*, *Varicorhinus lini*), and herbivorous fish (grass carp). Total mercury concentrations in fish ranged from 0.01 to 0.17 mg/kg wet weight, with a mean ( $\pm$  standard deviation [SD]) of  $0.04 \pm 0.03$  mg/kg wet weight (Fig. 5). The highest concentration was 0.17 mg/kg wet weight, observed in catfish, whereas the lowest concentration was 0.01 mg/kg wet weight, found in common carp. It is interesting to note that fish in newly built reservoirs in the Wujiang River contained low Hg levels, which is contrary to fish tissue from newly built reservoirs in North America and Europe. As shown in Table 5, Total Hg concentrations in catfish (*Parasilurus asotus*) from the HJD Reservoir were significantly lower than those of predatory fish species such as largemouth bass, pike, walleye, and perch collected from newly built reservoirs in Europe and North America. Meanwhile, THg concentrations in carp in the HJD Reservoir were significantly lower than those in nonpredatory fish species found in Europe and North America. No significantly linear correlation was observed between THg concentrations and body length or weight ( $p > 0.05$ ), as shown in Table 6, and THg concentrations in predatory fish were higher than those in nonpredatory fish, which indicated THg in fish was related mainly to the feeding habits of the fish and the trophic status. These results are consistent with a previous study of the Wujiang River [35], in which an average THg concentration of 0.06mg/kg wet weight was reported. It is generally accepted that more than 90% of the THg occurs in the form of MeHg in the edible portion of fish [36,37]. These concentrations were well below

Table 5. Comparison of total mercury concentrations (THg) in fish in Hongjiadu, China, with North America and Europe

| Sampling location                    | Periods | Fish species    |                   | THg (mg/kg)     | Data source  |
|--------------------------------------|---------|-----------------|-------------------|-----------------|--|
|                                      |         | Predatory fish  | Nonpredatory fish |                 |  |
| Keowee, Southeastern United States   | 5       | Largemouth bass |                   | 0.58–0.66       | Abernathy and Cumbie [2]                                   |
| Jocassee, Southeastern United States | 5       | Largemouth bass |                   | 1.9–4.9         | Abernathy and Cumbie [2]                                   |
| Southern Illinois, USA               | 5       | Largemouth bass |                   | 0.11–1.2        | Cox et al. [3]   |
| Northern Quebec, Canada              | 3–5     |                 | Longnose sucker   | 0.41–0.67       | Verdon et al.[5] Tremblay et al. [6] Schetagne et al. [42] |
|                                      |         |                 | Lake whitefish    | 0.43–0.58       |  |
|                                      |         | Northern pike   |                   | 1.2–2.8         |  |
|                                      |         | Walleye         |                   | 1.9–2.5         |  |
| Western and Northern Finland         | 3–6     | Pike            |                   | 1.6             | Porvari [7]  |
|                                      |         | Perch           |                   | 1.3             |  |
| Northern Manitoba, Canada            | 3–5     |                 | Lake whitefish    | 0.24–0.26       | Brouard et al.[4] Bodaly et al. [11]                       |
|                                      |         | Walleye         |                   | 0.73–0.78       |  |
|                                      |         | Northern pike   |                   | 0.70–1.1        |  |
| Southern Alberta, Canada             | 2       | Northern pike   |                   | 0.89            | Brinkmann and Rasmussen [43]                               |
| Hongjiadu Reservoir, China           | 5       | Catfish         |                   | $0.15 \pm 0.03$ | Present study  |
|                                      |         |                 | Crucian carp      | $0.10 \pm 0.04$ |  |
|                                      |         |                 | Common carp       | $0.03 \pm 0.01$ |  |
|                                      |         |                 | Grass carp        | 0.02            |  |

Table 6. Weight and length of fish in Hongjiadu Reservoir, China, and Pearson's correlation coefficients for the relation between total mercury concentrations (THg) and length/weight

| Species  | Number | Weight (kg) |                 | Length (cm) |               | Correlation coefficient |                |
|--|--------|-------------|-----------------|-------------|---------------|-------------------------|----------------|
|  |        | Range       | Mean $\pm$ SD   | Range       | Mean $\pm$ SD | THg vs. weight          | THg vs. length |
| Catfish ( <i>Parasilurus asotus</i> )          | 4      | 0.63–19     | 9.1 $\pm$ 6.8   | 45–95       | 71 $\pm$ 18   | 0.63*                   | 0.36*          |
| Common carp ( <i>Cyprinus carpio</i> )         | 30     | 0.30–1.5    | 0.50 $\pm$ 0.20 | 24–44       | 29 $\pm$ 3.6  | 0.84*                   | 0.65*          |
| Cruician carp ( <i>Carassius auratus</i> )     | 3      | 0.07–0.12   | 0.09 $\pm$ 0.03 | 15–18       | 16 $\pm$ 1.9  | 0.12*                   | 0.31*          |
| <i>Hemiculter leuciculis</i>                   | 23     | 0.01–0.05   | 0.10 $\pm$ 0.02 | 11–17       | 15 $\pm$ 1.6  | 0.80*                   | 0.56*          |
| <i>Varicorhinus lini</i>                       | 4      | 0.12–0.20   | 0.02 $\pm$ 0.01 | 21–25       | 23 $\pm$ 1.6  | 0.85*                   | 0.82*          |
| Grass carp ( <i>Ctenopharyngodon idellus</i> ) | 1      | 23,000      |                 | 91.3        |               |                         |                |

\*  $p > 0.05$ .

the national consumption limits for MeHg, which are 0.50 mg/kg for herbivorous fish and 1.00 mg/kg for predatory fish.

#### Possible reasons for low Hg levels in fish

As mentioned above, the reservoirs of the present study were deduced to be net sinks for THg and TMeHg lacking active sites for Hg methylation, in spite of the fact that THg concentration in precipitation and regional soil is elevated from an international perspective. It is generally believed that flooded soil is propitious for MeHg production [38]. Submerged soil in the relevant investigations from North America and Europe was normally from boreal forest or wetland, where organic matter (OM) concentrations can vary from 30 to 50% [10,15]. The high levels of OM have helped to create more favorable conditions for Hg methylation, which resulted in increased MeHg concentrations in the water columns and elevated MeHg concentrations in fish by bioaccumulation and biomagnifications through the food chain [12–15]. However, in the Wujinag River region, 72% of flooded areas are farmlands with reduced OM contents. Extensive agricultural activity in this region is the main cause of low OM concentrations in soil (Table 7). This is consistent with results from the State Environmental Protection Bureau of China (1990), China National Environmental Monitoring Center [27], and Liu [39], who reported that OM contents in soil ranged from 1.2 to 7.8% in southwestern China. It has been demonstrated that low MeHg concentrations in these reservoirs were associated with low OM in submerged soil. These results suggest that OM in soil is a key factor governing Hg methylation rates in reservoirs. It is obvious that further study is needed to elucidate the mechanisms of OM from soils for governing MeHg distribution in water column in both reservoirs.

Table 7. Comparison of organic matter (OM) levels in soil in the catchment of Hongjiadu and Suofengying reservoirs in southwestern China

| Sampling sites                     | OM%           | Source  |
|------------------------------------|---------------|---|
| Hongjiadu Reservoir, Guizhou       | 1.9 $\pm$ 1.1 | Present study                                       |
| Suofengying Reservoir, Guizhou     | 4.1 $\pm$ 1.3 | Present study                                       |
| Guizhou                            | 4.3 $\pm$ 2.7 | China National Environmental Monitoring Center [28] |
| Yunnan                             | 3.9 $\pm$ 2.4 |   |
| Sichuan (including Chongqing area) | 3.3 $\pm$ 4.5 |   |
| Tibet                              | 4.6 $\pm$ 4.7 |   |
| Southwestern China                 | 1.2–7.8%      | Liu [39]  |

Apart from OM levels of soil, MeHg production may be dependent on other physiochemical factors such as pH, DO, and watershed characteristics. In the present study, no obvious anoxic conditions were found in either reservoir, and pH levels were slightly alkaline (Table 1). It is well documented that the high pH and DO do not favor Hg methylation processes in aquatic systems, although the HJD and SFY reservoirs have different watershed areas, total water volumes, and water residence times (Table 1). Therefore, the environments of both reservoirs were not conducive to Hg methylation.

#### CONCLUSIONS

Newly constructed reservoirs are usually associated with MeHg pollution problems, but in the present study, THg and TMeHg concentrations in the water column and in fish tissue were much lower compared with those in North America and Europe. We suggest that this unexpected result is derived from the fact that the soils flooded by both reservoirs were initially characterized by low levels of OM, resulting in lower Hg methylation after flooding. Therefore, newly constructed reservoirs in southwestern China may not have significant MeHg pollution.

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